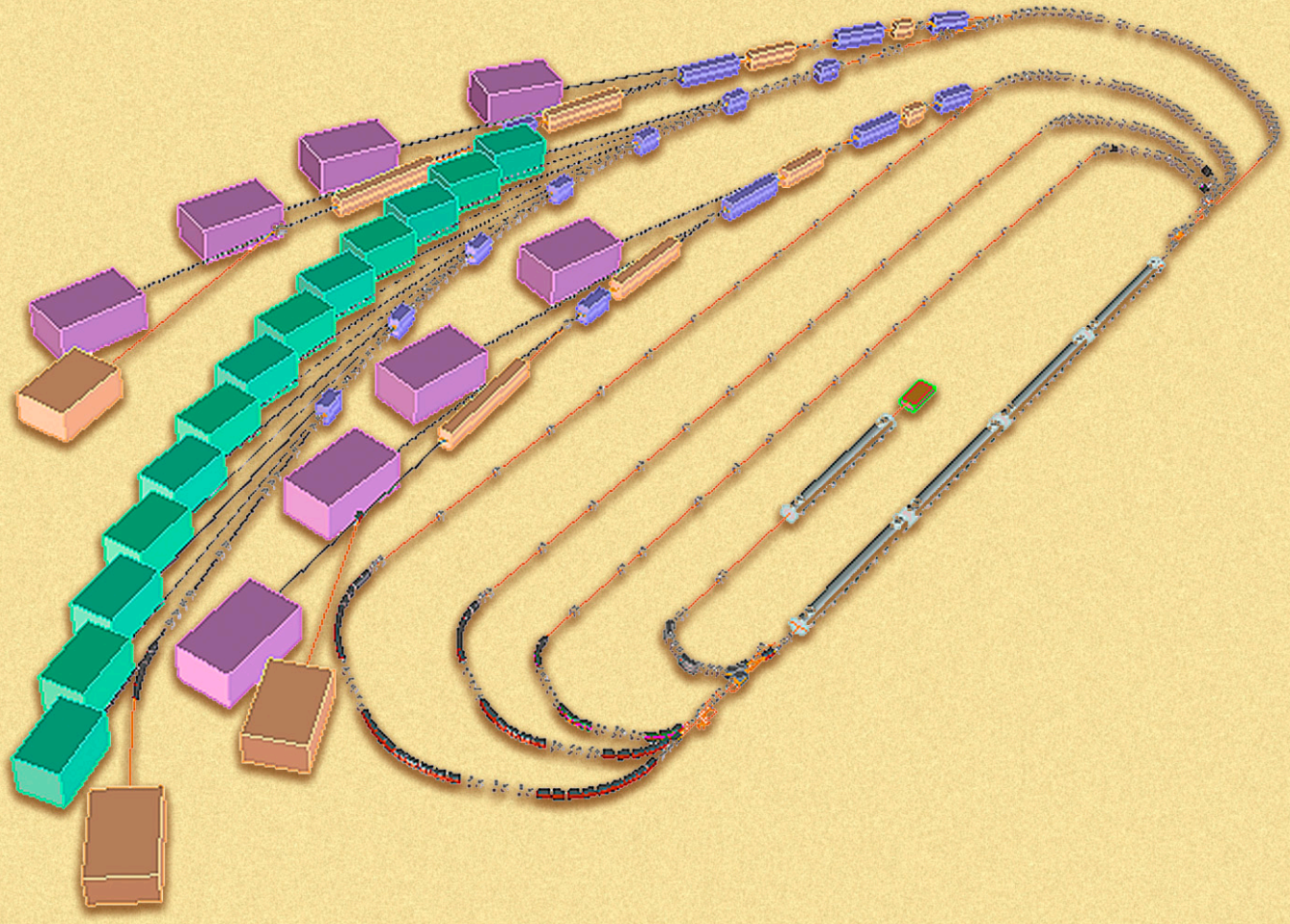
LUX

A Linac-Based Ultrafast X-Ray Source for the Future



September 2003
Berkeley, California

LBNL's Vision for a National Emphasis on Ultrafast X-Ray Science

- Vision for ultrafast science – why lasers have played an enormous role in new science that has been done
- Ultrafast studies have been crucial in elucidating fundamental processes of vision, photosynthesis, semiconductor processes and semiconductor lasers, and conversion of light to other forms of usable energy
- The ultrafast time domain encompasses 100 femtoseconds down to attoseconds – the timescales of atomic motions
- There is a growing national and international interest in ultrafast processes that can be performed with x-rays – this interest cuts across all fields of science
- Lasers perform all of the ultrafast experiments we know of today, but they are unlikely to produce substantial fluxes of x-rays of 0.1-12 keV in the future
- The time-energy uncertainty principle favors the ability to achieve significant spectral resolution in the x-ray region, while still producing ultrashort timescales – chemical and structural specificity possible
- The x-ray region is where attosecond time dynamics will be achieved – this is a key driver for the vision of the lab
- In the visible, the bandwidth of attosecond pulses would be proportionately so large that no spectral or state resolution is possible
- Major new directions in science with ultrafast x-rays include the direct obsevation of chemical transition states, dynamics on the timescales of electron motions, nonreversible modifications of biological matter, magnetic effects induced by merging magnetic materials with semiconductors, interfacial phenomena, and molecular wires and molecular electronics
- Electronic dynamics will be a key new area of major impact
- Almost no ultrafast diffraction experiments have been possible, and structural information on ultrafast timescales will address many needs
- X-ray spectroscopies have markedly different features compared to visible, infrared, and ultraviolet spectroscopies, and can reveal important structural information – other novel spectroscopies are already anticipated in theoretical publications
- Accelerator-based approaches using highly refined, high energy electron beams have the best chance to produce high-flux ultrafast x-rays across the whole wavelength spectrum
- 40 light sources worldwide – none emphasize time domain dynamics – opportunity for natural ties between ALS and an ultrafast x-ray facility at LBNL
- LBNL has developed a concept to produce ultrashort x-ray pulses as a user facility and in a highly refined manner for experiments across all fields
- There are many competing technologies being developed worldwide
- The recirculating linac concept starts with relatively short bunches of electrons compared to synchrotrons – by careful design, the electron beam quality can be maintained with 2 ps bunch duration
- Short pulse laser seeding and x-ray pulse compression methods permit these short electron bunches to produce substantial fluxes of x-rays as short as 10 femtosecond duration

- There are excellent prospects to achieve attosecond pulses in the future
- The facility is designed from the beginning to be synchronized with ultrafast lasers, so that pump-probe and more complex multidimensional spectroscopies can be performed
- Lasers are a major integral component of LBNL's concept facility to perform experiments - by initiating photoprocesses in samples and then probing them with x-ray spectroscopies and diffraction
- Positive comments from BESAC sub-committee on 20-year Facilities Roadmap:
  - "This is an innovative proposal for an ultrafast X-ray and VUV facility"
  - "The proposal presents a strong scientific case, based on a large community involved in femtosecond chemistry and physics who could take advantage of this source"
  - "The BESAC Subcommittee applauds this group for its vision and innovation"
- BESAC report recommendations for LBNL's recirculating linac-based facility:
  - Need for national reviews of science case, national competition for new facility
  - Consider high cost
  - Potential weakness of low average power
  - Cascaded HGHG not demonstrated
- We are addressing these, and other, concerns:
  - Identified needs for accelerator, insertion device, and detector R&D and science case
  - Clearly define differences with LCLS – synchronization, refined tunability for experiments across all disciplines of science
  - Management plan and relationship to ALS – (1) continuous and (2) time-dynamic x-ray spectroscopy facilities
- Strengthen scientific case and build national support:
  - Napa workshop organized by LBNL, Montreux, Switzerland workshop, Corsica Summer school
  - Many broad categories of important experiments and detailed examples identified
  - Convene mini-working groups to brainstorm science
  - Workshop FY'04 to discuss science with a recirculating linac ultrafast facility
- Address technical questions with R&D
  - Partner with BNL to test harmonic generation in FEL's over two stages

LUX Facility Performance Specifications

- Soft X-Rays
  - 20 eV – 1 keV
  - 10 fs – 200 fs
  - 10^{10}-10^{13} ph/pulse/0.1%BW
  - Variable polarization
  - 10 kHz repetition rate
  - Spatial and temporal coherence

- Hard X-Rays
  - 1 keV – 12 keV
  - 50 fs – 100 fs
  - 10^{6} ph/pulse/0.1%BW
  - Linear polarization
  - 10 kHz repetition rate
  - Spontaneous emission

- Synchronization
  - 20 fs soft x-ray
  - 50 fs hard x-ray

- Laser systems
  - Laser oscillator serves as facility master oscillator
  - Stabilized all-optical timing distribution system
  - 267 nm photocathode laser
  - 200-150 nm tunable seed for soft x-ray production in FEL's
  - Multiple tunable 267-3000 nm experiment initiation laser systems
    - Stable and controlled environment at each endstation
    - Optical manipulation and diagnostics

LUX - Linac-based Ultrafast X-Ray Source

A Recirculating Linac/Laser-based Femtosecond Facility for Ultrafast Science

Overview

A recirculating linac user facility is proposed to address the growing national and international need for ultrafast x-ray scientific research. The LUX facility is based on recirculating linear accelerator technology, coupled with an array of advanced tunable femtosecond lasers, and is capable of performing an enormous variety of pump-probe type experiments with soft and hard x-rays. The facility has been specifically designed with a view toward solving problems in ultrafast science, and it's impact will be across all fields of science, from biology, chemistry, and physics, to novel areas such as quantum computing, spintronics, and highly nonlinear phenomena. LUX follows a decade of ultrafast facility development at LBNL, and represents the next stage of development following the ALS laser-slicing source currently under construction. The science case for an ultrafast x-ray facility has developed over this timescale and is maturing as the ALS slicing source nears operational status.

The recirculating linac accelerates picosecond-duration electron bunches to 2.5-3 GeV. Intense soft x-rays are produced by cascaded harmonic generation scheme, similar to high-gain harmonic-generation (HGHG) - a laser-seeded process in a cascaded series of free-electron lasers (FEL's), resulting in enhanced radiation at selected harmonics of the seed. The coherent soft x-rays can be tuned over a range of tens of eV to 1 keV, and ultrashort seed laser pulses produce pulse durations of 10-200 fs. Hard x-rays are produced by spontaneous emission of the electrons in narrow-gap, short-period undulators. By use of a novel bunch tilting process followed by optical compression, hard x-ray pulse durations of 50-100 fs are obtained over a range of 1-12 keV. Synchronization of the x-rays with lasers is critical for experiments, and optical pulses initiate both harmonic generation seed lasers and experimental end station amplifiers for precise timing. The femtosecond x-rays are produced at a 10 kHz repetition rate, with variable polarization, and with peak fluxes comparable to third generation light sources. With these capabilities, the LUX facility allows unprecedented studies of time domain processes with x-rays.

The facility has capacity for approximately 20 endstation bays, each with integral laser systems for pump-probe experiments, and an initial complement of 8 beamlines is sought. Multiple user groups may set up at a given beamline, running in shifts, and approximately 60 user groups are accommodated round the clock for 2-3 month periods necessary to perform challenging new time-resolved x-ray science experiments in magnetism, spintronics, structural biology, phase transitions, soft condensed matter, and chemical dynamics. The LUX facility has flexibility for upgrades, when experiments require higher fluxes and shorter pulses. An energy recovery option can be introduced for higher average beam powers. An FEL slicing technique with potential for high energy attosecond pulses is being developed, and the facility is well positioned to remain the major contributor to ultrafast x-ray dynamics in the world for many years to come.

From the many exemplary studies already presented at workshops, a substantial grass roots effort to begin the field of ultrafast x-ray science has already occurred, and there is no doubt that the LUX facility will be in high demand and oversubscribed. The facility will be unique among DOE ventures, because it marries ultrafast time-domain measurements with x-ray science. The whole spectrum of already-available x-ray determinations, long a staple at synchrotron facilities, and vigorously producing results in all fields of science, would be open to time-dependent measurements. The unique design of the LBNL facility will satisfy an enormous range of pulse properties for each area of specialty. There will be outstanding synergism with other facilities such as the Advanced Light Source (ALS) and the Molecular Foundry. The x-ray pulse slicing beamline currently under development at the ALS, and capabilities in nanoscience at LBNL, will merge directly into the important science accomplished at LUX, and the facility will become an international attraction for time-dependent measurements. The extensive expertise in ultrafast laser measurements and laser development in the San Francisco Bay Area ensures a high likelihood of finding solutions of unique and challenging scientific problems, and an exciting environment for students to flourish in an emerging field. The facility will be as much a laser-related set of tools as an accelerator system, with world-renowned expertise in how to produce and use ultrafast lasers and x-rays to solve real scientific problems. We anticipate that advances in lasers in the next decade will be remarkable, and these advancements will also represent continual upgrades to the facility, both through seeding and for direct excitation at end stations, even without major changes to the accelerator structure.

The importance of the science

Ultrafast x-rays have been identified world-wide in numerous workshops and reports as a key area ripe for new scientific investigations. Lasers successfully cover most of the visible, infrared, and ultraviolet regions of the spectrum with both high resolution and very short pulses. Thus, experimentalists have utilized lasers to tremendous advantage for thousands of time-dynamics investigations, many absolutely critical to the scientific fields of solid state physics, semiconductors, photochemistry, and photobiology. Until now, ultrafast time domain studies in the x-ray region have been almost completely lacking. By use of synchrotron radiation and by novel conversion of intense laser pulses into soft and hard x-rays, scientists have been able to perform some of the first innovative experiments recently, such as Bragg diffraction studies of phase transitions, time-resolved Laue diffraction of myoglobin-CO reversible binding, femtosecond photoelectron spectroscopy, and even attosecond electron redistribution in Auger electron processes. However, these laser-based x-ray fluxes are low, the signal levels weak, and experiments are challenging to accomplish by individual scientists. The LUX recirculating linac-based facility proposed here provides an increase of x-ray flux by several orders of magnitude, is accessible to a large number of users, with resources available for set-up of pump-probe femtosecond-scale time resolved experiments utilizing ultrafast lasers.

While the approximately 40 available light sources in the world are largely limited to static spectroscopies, microscopies, and structures, this facility will be the first designed from the start as a user facility for femtosecond x-ray dynamics, with precise timing as an integral requirement. The LCLS has the potential to demonstrate some of the first exciting ultrafast x-ray studies with an accelerator-based machine. LUX will be a highly refined ultrafast x-ray source, offering higher repetition rates but lower pulse energies than LCLS, tunability, and precision timing with other laser sources for excitation and probe experiments. It will accommodate many users at one time across the whole spectrum of experimental possibilities. The science to be carried out with the LUX facility cuts across all scientific disciplines.

By combining both diffraction to explore nuclear positions in real time and spectroscopy to interrogate electronic and atomic states and their structural parameters and chemical environments, the facility represents a powerful combination to address scientific problems. Broad categories of possible experiments include:

- Photoinduced phase transitions
- Metal-insulator photo-induced transition
- Magnetics, photon-excited ferromagnetism, spintronics
- Time domain structural biology
- Solute-solvent structural dynamics and charge switching
- Surface transformations
- Laser-induced continuum dressing of atoms & molecules
- Nanoparticle physics
- Plasma physics
- Liquid microjet photochemistry studies
- Interfacial phenomena
- Soft condensed matter, time-domain microscopy

And the basic techniques of interrogation involve:

- Pump-probe with visible laser pump light
- Twin x-ray pulses, one and two color
- Coherence and multidimensional spectroscopies, four wave mixing, x-ray probe
- X-ray near edge absorption spectroscopy
- Photoelectron spectroscopy
- Photoemission microscopy
- X-ray magnetic dichroism
- Time-resolved Laue diffraction
- Magnetic speckle
- Scanning transmission microscopy
- Time domain XPS

Although pump-probe experiments represent some of the most important techniques, involving a femtosecond laser as a pump and the ultrafast linac-based x-ray source as the probe, the facility will also be designed to accommodate rapidly emerging multidimensional coherent laser spectroscopies (e.g. three-laser pump beams and an x-ray probe), as well as two x-ray wavelengths, for double-resonance x-ray pump and probe spectroscopies. Most of these novel forms of spectroscopies with x-rays have not even been delineated yet. The ability to perform high resolution near edge x-ray spectroscopy, magnetic dichroism, time domain speckle, and time-resolved Laue diffraction are critical. Sample damage is kept to a minimum with lower energies and higher repetition rates, in many cases with complete sample regeneration by translation or flow.

Outline of the facility

Feasibility studies carried out at LBNL show that the proposed facility is feasible and can be built with existing technology, with engineering developments for this particular application. These studies have explored a variety of machine concepts in pursuit of a facility to meet the needs of x-ray studies of ultrafast dynamic processes, leading to the recirculating linac design for LUX. The major components and systems of LUX are already known accelerator

technologies: an rf photo-injector, superconducting linear accelerators, magnetic arcs and straight sections, pulsed extraction magnets, transversely deflecting cavities, cascaded harmonic-generation, narrow-gap short-period undulators, x-ray manipulation in optical beamlines, and a variety of short-pulse laser systems. The flexibility of the LUX lattice design allows control and preservation of electron beam transverse and longitudinal emittances, minimizing the influence of collective effects. The machine layout is shown in Figure 1. After acceleration to 2.5-3 GeV, the electron bunches pass through undulators to produce radiation over a range of a few tens of eV to 12 keV. EUV and soft x-rays are generated by a seeded free-electron laser process in a cascaded harmonic-generation scheme (similar to HGHG). Hard x-rays are produced by spontaneous emission of the high energy electrons in short-period undulators.

The physics and engineering design has been developed sufficiently to confirm the feasibility of the project, and to produce a preliminary costs estimate. LUX is based on accelerator technologies that have already been successfully demonstrated on operating machines. We have identified engineering development requirements, and developed risk mitigation plans with an R&D program. Significant R&D has already been invested in the accelerator technologies required, and is ongoing in many areas that may be expected to lead to advances beyond our baseline design parameters, and thus to improved future performance.

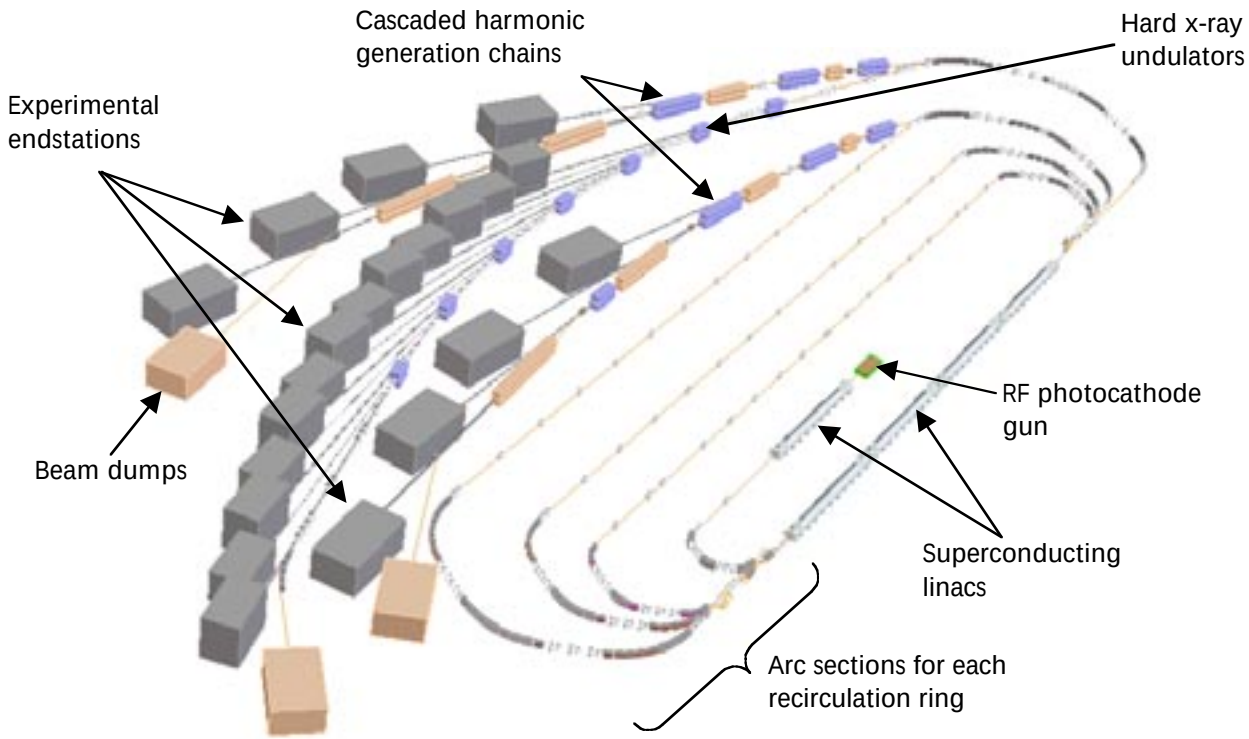
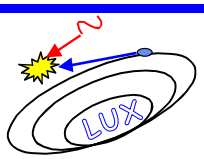


Figure 1. Machine layout showing experimental beamlines cascaded harmonic-generation chains, and major accelerator components. The machine footprint is approximately 150x50m. A capacity of approximately 20 beamlines is shown, an initial complement of 8 is proposed.



LUX - Linac-based Ultrafast X-Ray Source

Scientific need

- Proposal based on growing national and international interest in ultrafast processes that can be performed with x-rays
  - Cuts across all fields
    - Napa, Montreux, Corsica workshops
- Ultrafast encompasses 100 femtoseconds down to attoseconds, where timescales of most nuclear motions are relevant
- Time-energy uncertainty principle favors the ability to achieve significant spectral resolution in the x-ray region, while still producing ultrashort timescales
  - Chemical and structural specificity possible

Scientific need, con't.

- The x-ray region is where attosecond time dynamics will be achieved, electronic motions are relevant
  - In the visible, the bandwidth would be so great that spectral resolution is not possible
- X-ray spectroscopies have markedly different features compared to visible, IR, and UV, and can reveal important structural information
- Almost no ultrafast diffraction experiments have been possible, and structural information on ultrafast timescales will address many needs
- Electronic dynamics will be a key area of major impact

Facility concept

- Lasers perform all of the ultrafast experiments we know of today, but they are unlikely to produce substantial fluxes of x-rays of 1-10 keV in the future
- LUX is a concept to produce ultrashort x-ray pulses as a user facility and in a highly refined manner for experiments across all fields
- The recirculating linac concept starts with much shorter bunches of electrons compared to synchrotrons
  - By careful design, the electron beam quality can be maintained with ~ 2 ps bunch duration

Facility concept, con't.

- Short pulse laser seeding and pulse recompression methods permit substantial fluxes of x-rays of at ~ 10-200 fs duration
  - Prospects for even shorter pulses
- The facility is designed from the beginning to be synchronized with ultrafast lasers
  - Pump-probe and more complex multidimensional spectroscopies possible
- Lasers are a major integral component of the facility to perform experiments
  - Initiating photoprocesses in samples and probing them with x-ray spectroscopies and diffraction

Facility concept, con't.

- Combine management and service operations of LUX and ALS, while at the same time taking advantage of large user base at ALS for novel time domain experiments
- Combined facility provides major thrust in experiments with x-rays both
  - (1) continuous
  - (2) time dynamics

Specifications of the facility

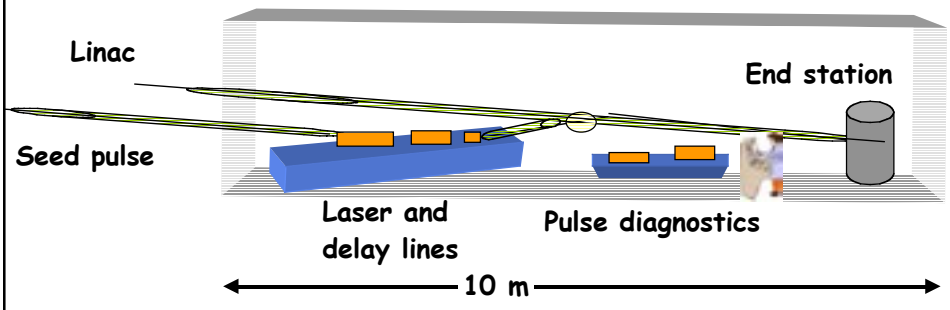
An ultrafast x-ray science user facility addressing scientific needs in Physics, Chemistry and Biology

- National user facility
  - recirculating linac-based light source
  - multiple beamlines
  - laser-coupled end stations

• Repetition rate	10 kHz
• Synchronization	~ 10's fs
• Pulse durations	10-200 fs, <1 fs in future
• Polarization	fully variable
• Broad photon range	~ 0.02 - 12 keV
• Photons per pulse	10^7 hard x-ray, 10^8-10^{12} soft x-ray

Typical end station layout

- Precisely timed laser and linac pulses



- Tunable laser systems designed for specific experiments, repetition rate, energies

Parameters that matter most to ultrafast scientists

Searching for weak dynamically changing signals in the midst of large time-invariant signals

- **Timing**

  most experiments performed by initiating a time evolving process with another laser or x-ray pulse - tight time synchronization is expected
  - 10's of fs for seeded, 50 fs for hard x-rays
- **Pulse-to-pulse stability**

  essential since only a small fraction of the molecules or materials are excited
  - Expectation of 3rd generation 0.1% stability, Real time subtraction - pump on/pump off
- **Bandwidth and chirp**

  BW a minimum, without violating transform limit, to isolate spectral shifts, chirp to correlate energy with time in new ways
  - Core level shifts of 0.1-0.5 eV typical, NEXAFS ≤0.1 eV desirable

Con't

Con't

Ultrafast scientists' needs

- **Tunability**
  - Spectroscopy demands tuning to near edge transitions
- **Polarization**
  - Complete rhc and lhc components needed for polarization blocking and dichroism experiments
- **Repetition rate**
  - High repetition rates desirable for samples that can be refreshed, low damage, as high as conventional electronics
- **Pulse duration**
  - 50-200 fs for many processes, 10 fs for some applications, 100 attosecond beyond
- **Pulse energies**
  - Sufficient pulse energies to obtain photoemission signals, absorption contrast changes, without sample damage
- **Coherence**
  - Spatial and temporal, speckle experiments

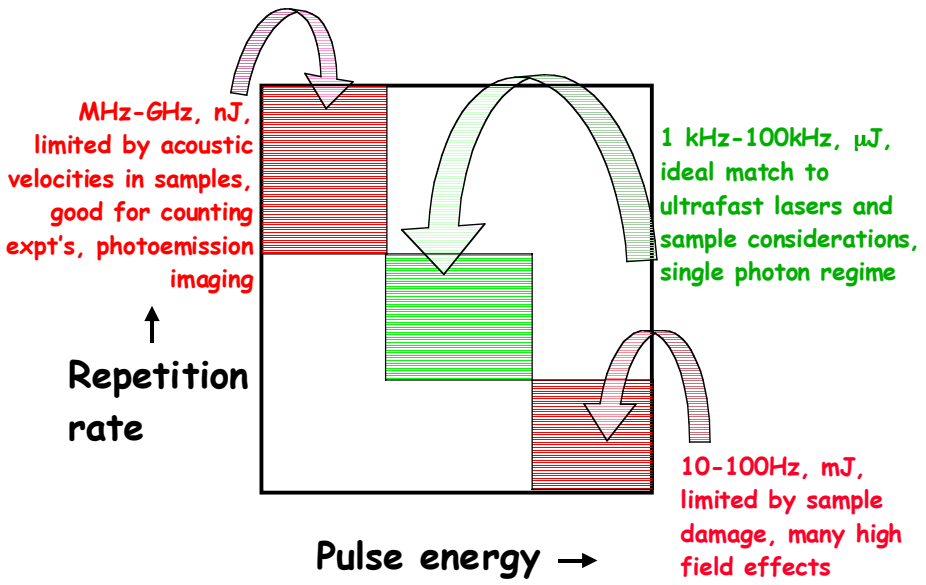
Con't

Ultrafast scientists' needs

- Focusability
  - Near-diffraction limit for seeded systems, 10's nm at 1 keV
- Power density
  - 10^{15} W/cm^2 readily achievable
- Trade off between power density and repetition rate
  - Maintain linear probing for many experiments
  - Multiphoton versus single photon

Recirculating linac design gives best opportunity to achieve experimental and facility goals

Repetition rate vs. energy



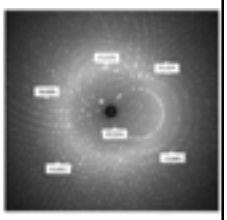
The fs linac science case

- Why unique and important - National and international user base - young scientists interested in ultrafast processes, tremendous grass roots efforts growing in ultrafast x-ray science - answer critical questions with national need

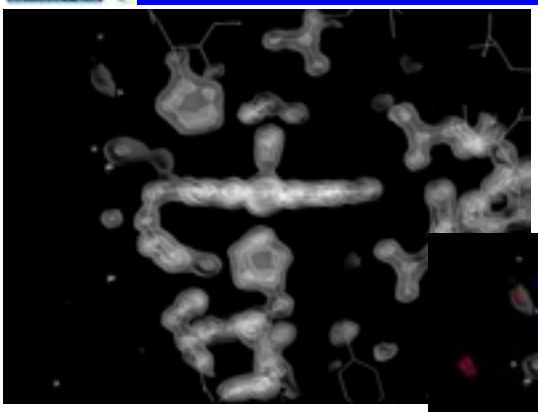
  - recirculating linac design is fundamentally a different source from SASE process
  - an excellent,highly refined platform for a user facility
  - allows major national thrust for time-dynamics investigations in the x-ray
  - timing and synchronization, matched to laser excitation sources, and laser seeding, are central concepts for success

Biomolecule crystallography

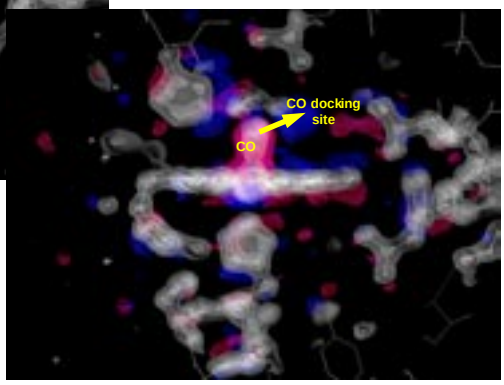
- Key time-resolved x-ray Laue diffraction experiments demonstrated at ESRF on photoactive systems, single crystal myoglobin and yellow protein
- Some biological processes inherently slow, others impressively fast, and these will become targets of investigation
- More systems will be developed and explored on ultrafast timescales as fs x-ray sources become available
- Probe desirable photochemically active, reversible sites related to plant photochemistry and vision, as well as the time-resolved nonreversible photon and particle damage of biologically active materials, related to cancer and mutations



Biological structural studies



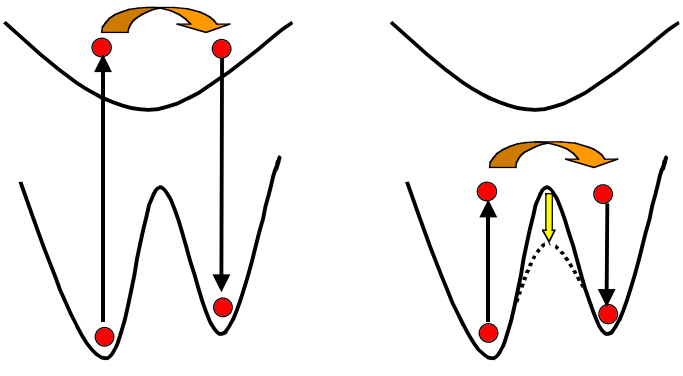
- ESRF ID09 10^7 photon per pulse per 0.1% BW
- LUX is comparable, with higher repetition rate



3 ns after optical pulse

Solid state phase transformations

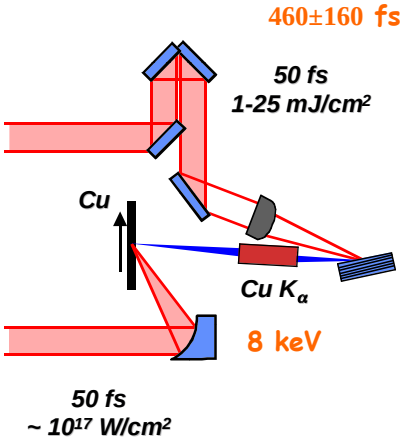
- Multiplicity of mechanisms for phase transitions

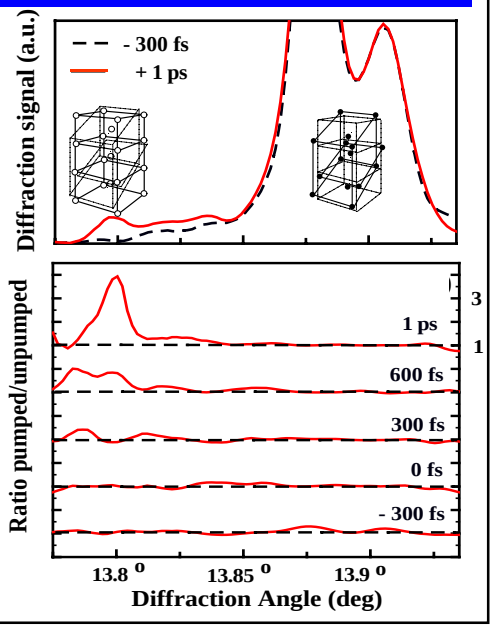


photoinduced

thermal activation

Bragg diffraction



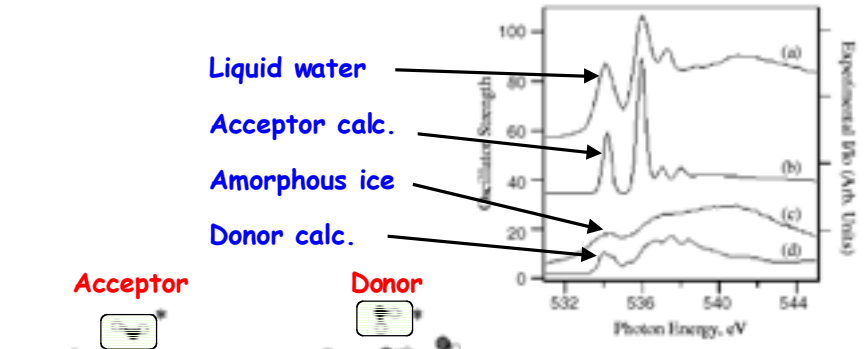


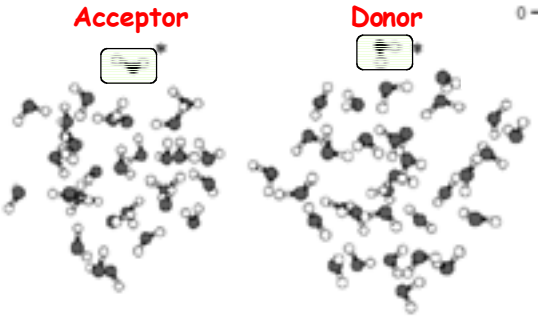
- Only 2000 Cu K_α photons per pulse, 20 Hz, only topmost layers altered

Liquid microjet studies of surface structural changes on ultrafast timescales

- An example of an experiment that requires:
  - Narrow Bandwidth
  - Pulse-to-pulse stability
  - Tunability
- Liquid water molecules
  - Probe O atom K edge at 530 eV
  - Obtain structure and bonding of surface water molecules
  - Sensitively probed by short escape depth of ions
- New ultrafast experiments
  - Excite Surface molecules vibrationally and photolytically and observe surface structural changes, time domain interfacial studies, caging
  - Hydrocarbon, salt and alcohol dopants segregate at surface, rich chemistry
- Advantages
  - Complete sample regeneration
  - Power densities limited primarily by space charge, affects imaging

Experimental NEXAFS of Liquid Water Surface

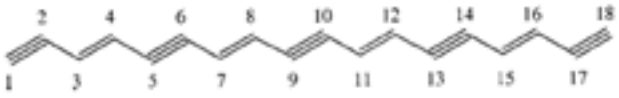


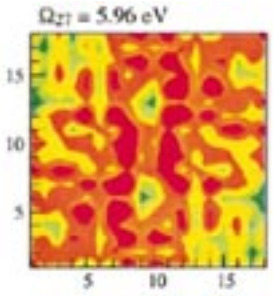


- Hydrogen-bonded donor and acceptor
- Acceptor structure fits better

Novel limits of short pulse x-rays

- Time-resolved x-ray Raman spectroscopy
- An inelastic Raman scattering of photons that reveals valence electron states
- Novel probe of electronic and vibrational motions in time
- Signals are shifted in energy a few eV from the large elastically scattered photon flux

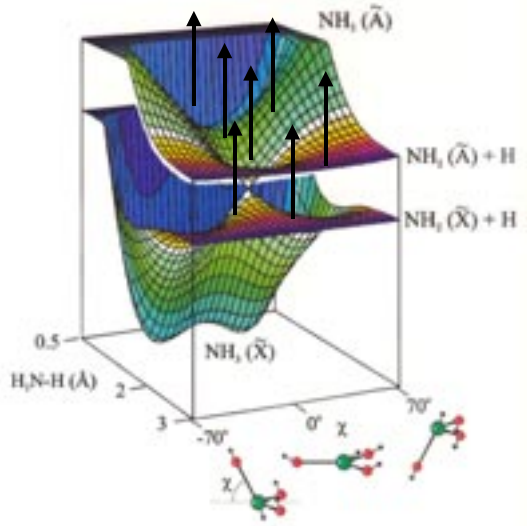




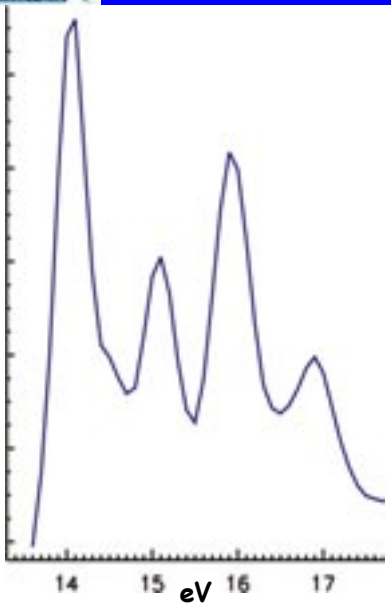
- Tanaka, Volkov, Mukamel - polydiacetylene
- Transition density matrix for 27th excited state vs. carbon atom position

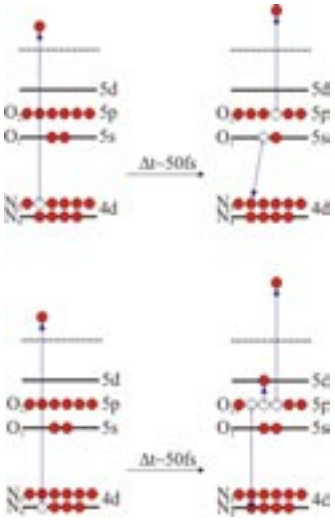
Atom specific probing of transition states in real time

- Probing transition states in real time



"Femtosecond" Auger spectroscopy

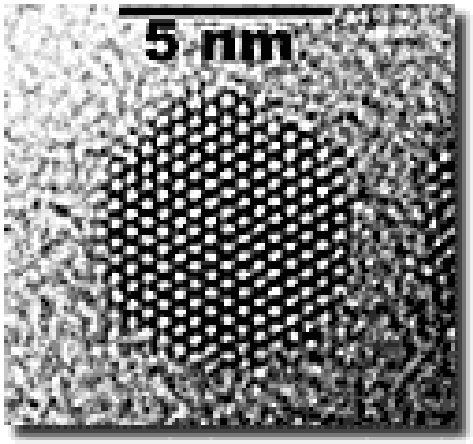




- "dream" exp'ts on sub-fs timescales

Quantum dots, superposition states and quantum information

- Semiconductor quantum dots - probing localized charge distributions, confinement, and exciton entanglement



Recirculating linac provides a platform for a refined source of ultrafast x-ray pulses

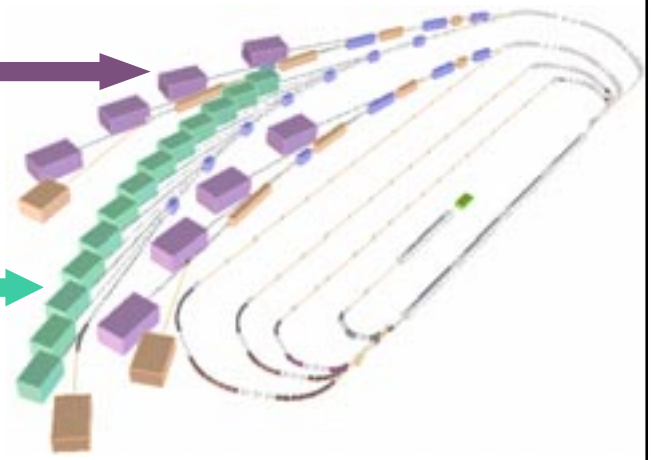
- RF photocathode gun produces high-quality electron beam
  - 3 mm-mrad, 1 nC, 20 ps
- Ability to produce "flat" beam with small vertical emittance
- Compress 120 MeV beam to 2-3 ps
- Accelerate in multiple passes through linac
- 2.5-3 GeV beam generates x-rays
  - 10-100 fs duration
- Compact (~ 150 x 50 m)
- Flexible configuration
  - Each pass provides opportunities for
    - Manipulation of the electron beam
    - Photon production
  - Variable repetition rate
  - Energy recovery option

LUX provides a wide range of x-ray wavelengths, operating simultaneously

- Tuneable x-ray beams from undulators

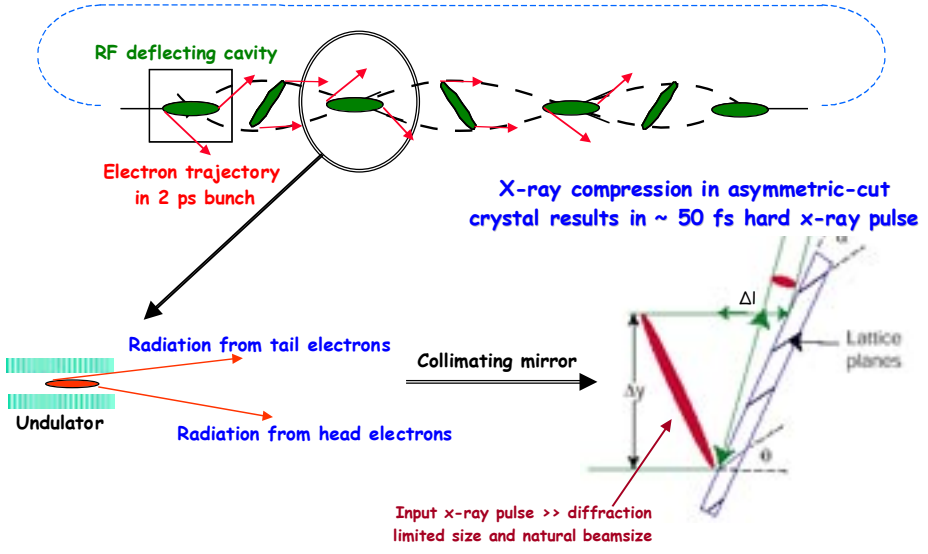
- Soft x-rays
- Laser-seeded cascaded harmonic-generation
  - 20-1000 eV
  - Spatial and temporal coherence
  - 10-100 fs

- Hard x-rays
- Spontaneous emission in narrow-gap short-period insertion devices
  - 1-12 keV
  - 50-100 fs



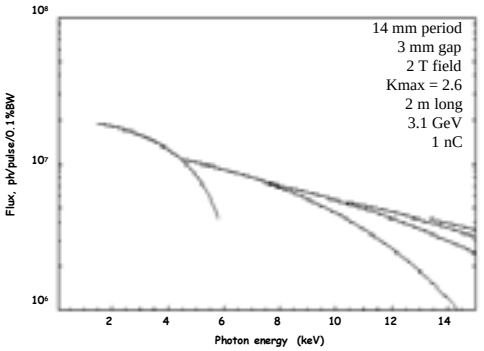
Hard x-ray pulses by electron bunch manipulation and x-ray compression

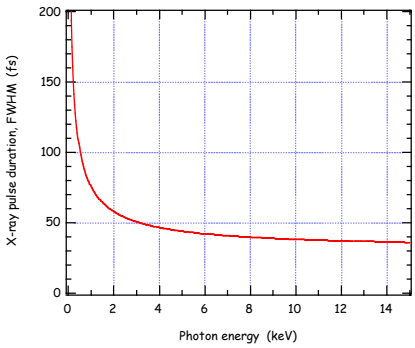
Conceived by A. Zholents, LBNL [1]



[1] A. Zholents et al "Generation of subpicosecond x-ray pulses using RF orbit deflection", NIM A 425 (1999) 385-389

LUX - hard x-ray pulses

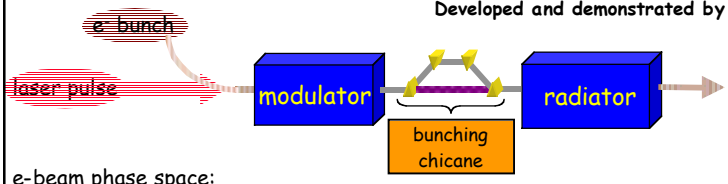




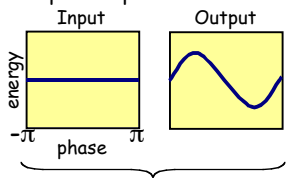
- **Same flux/pulse as 3rd generation light sources**
- **1000 times shorter pulse**
  - **Beam emittance dominates at shorter wavelengths**
    - **Need for low emittance electron beam**
  - **Optical diffraction dominates at longer wavelengths**

Harmonic generation scheme

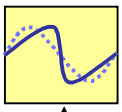
Developed and demonstrated by L.-H. Yu et al, BNL [1]



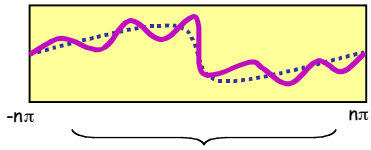
e-beam phase space:



Energy-modulate e-beam in undulator via FEL resonance with coherent input radiation



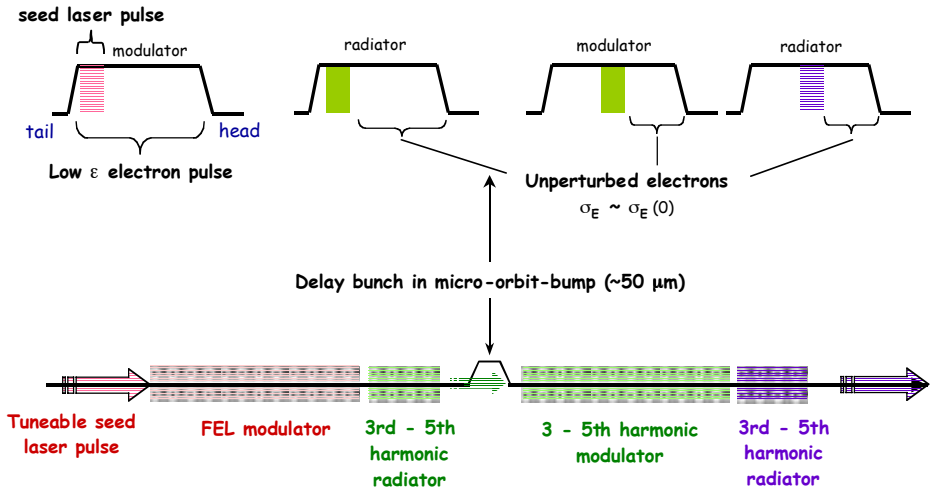
Dispersive section strongly increases bunching at fundamental wavelength and at higher harmonics



In a downstream undulator resonant at λ_0/n, bunched beam strongly radiates at harmonic via coherent spontaneous emission

[1] L.-H. Yu et al, "High-Gain Harmonic-Generation Free-Electron Laser", Science **289** 932-934 (2000)

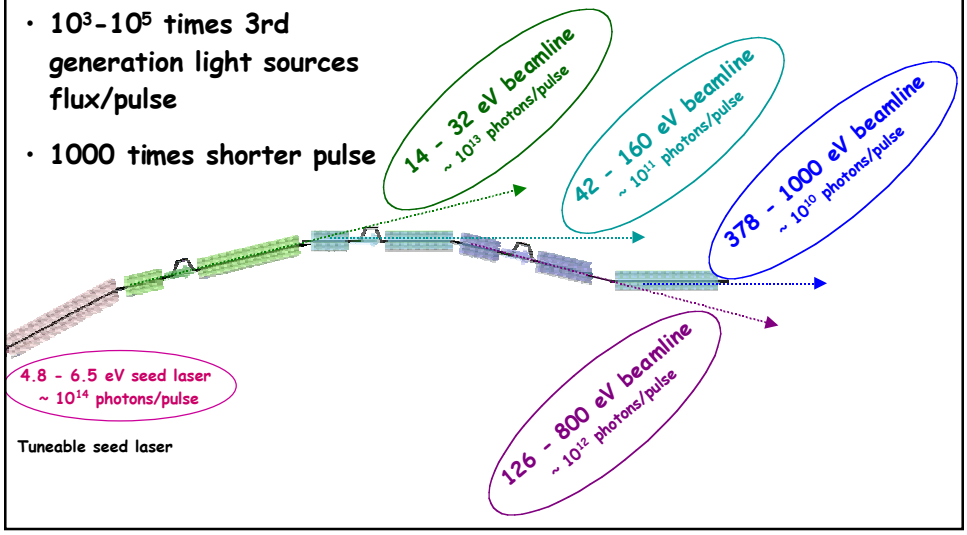
Cascaded harmonic generation scheme



- 4 stages produces beamline with energies ranging from ~ 20 eV to 1 keV

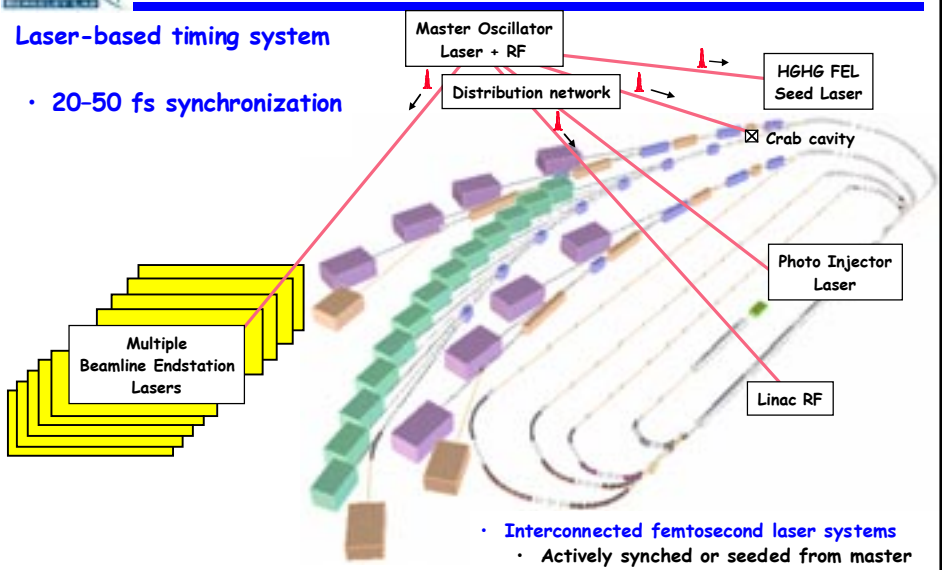
LUX - multiple beamlines with 4-stage harmonic generation

- Wide range of soft x-ray wavelengths accessible by tuning seed OPA and undulators
- 10^3-10^5 times 3rd generation light sources flux/pulse
- 1000 times shorter pulse



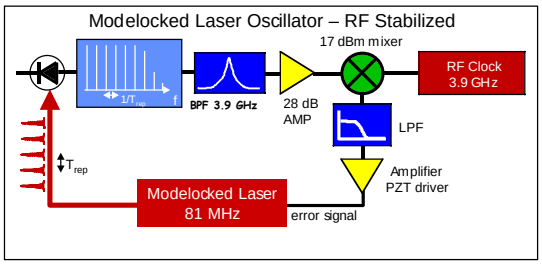
LUX facility is designed to achieve synchronization between pump and probe

- Laser-based timing system
  - 20–50 fs synchronization

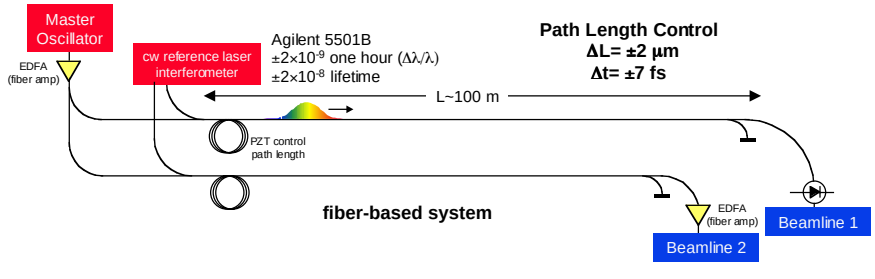


- Interconnected femtosecond laser systems
  - Actively synched or seeded from master
  - Laser to laser synchronization
  - Stabilized timing distribution network

Master oscillator and timing distribution

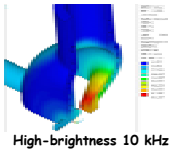


- Phase-lock all lasers
- Derive rf signals from laser oscillator
- Rf feedback of deflecting cavities
- Synchronization of 20-50 fs

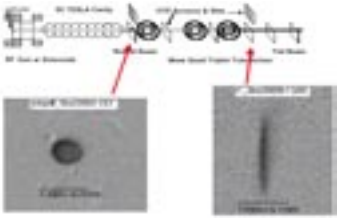


Technologies for LUX exist, proposed engineering and physics developments will meet requirements

- High-brightness high-rep-rate photocathode gun
  - Required 3 mm-mrad @ 1 nC demonstrated, we have developed high-power design
- Flat-beam production
  - <1 mm-mrad demonstrated, we are collaborators in this experiment
- CW superconducting RF
  - We have developed engineering modifications for the TESLA design, TJNAF upgrade may use 20 MV/m
- Lasers and optical distribution
  - We are developing laser expertise at existing ultrafast experiments (ALS, L'Oasis)
- Superconducting narrow-gap undulators
  - We are developing designs and harmonic correction schemes, Karlsruhe & ACCEL work
- Cascaded harmonic generation
  - We are planning activities to demonstrate staged harmonic generation



High-brightness 10 kHz RF photocathode gun

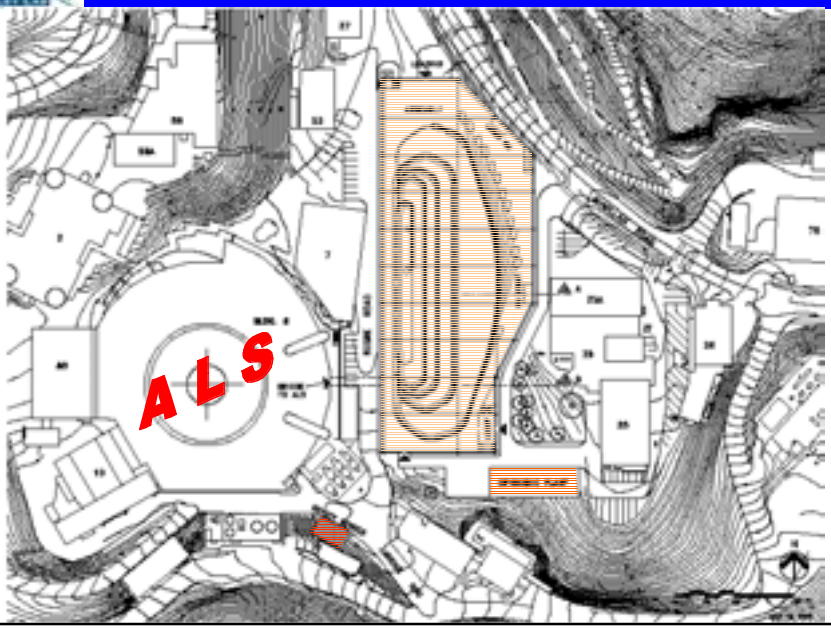


Round beam image on fluorescent screen

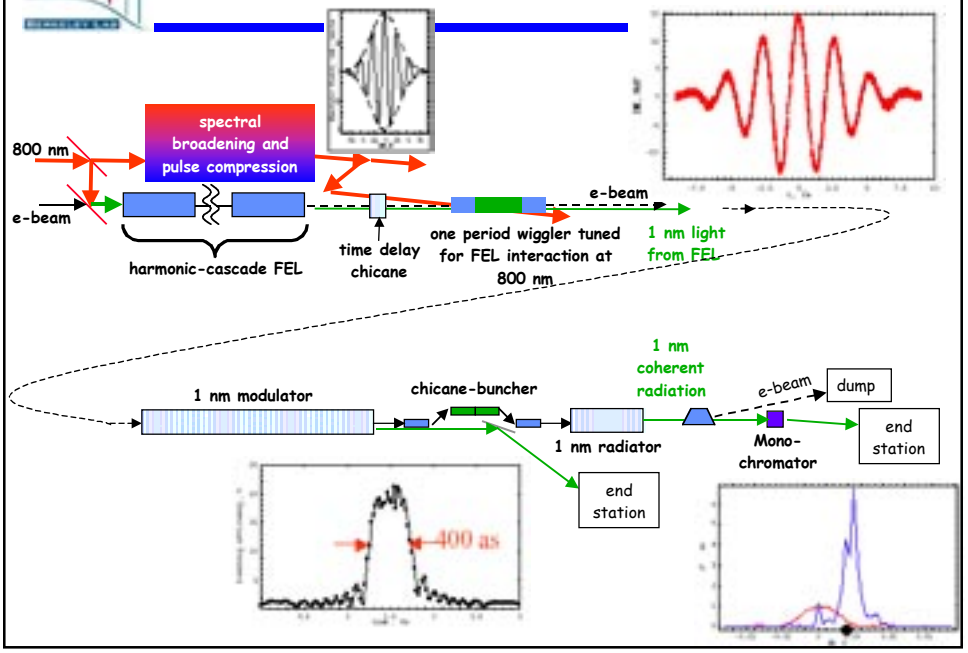
Flat beam image on fluorescent screen

J-Lab upgrade cavity

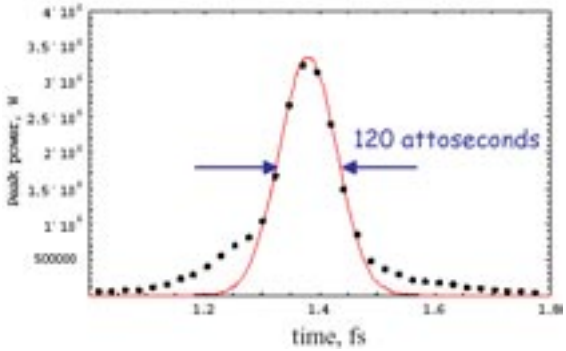
Several potential sites identified "Old Town" site maximizes synergies with ALS



Schematic of attosecond production



Attosecond radiation at 1nm wavelength



A snap-shot of the radiation field at the end of the 2m long radiator-undulator.

Dots show the result of the simulation and red line shows the Gaussian fit.

Science with Soft X-Rays

"But, soft! What light ... "
Romeo and Juliet

High brightness synchrotron radiation in the soft x-ray and ultraviolet regions offers improved spectral resolving power, spatial resolution, and coherence. Examples are presented of science being done in the areas of wet cell biology, condensed matter physics and extreme ultraviolet optics technology.

Neville Smith

Introduction

Users of synchrotron radiation fall into two fairly distinct camps: users of the soft x-ray and vacuum ultraviolet (VUV) region of the spectrum; and users of the hard x-ray region. The distinction can be expressed quantitatively by comparing the energies of a photon and an electron whose wavelengths are one Ångstrom. The scientific question posed by the users of hard x-rays tends to be "where are the atoms?" with much emphasis on the determination of crystal structures and molecular structures using techniques such as x-ray diffraction. The probing *photon* should therefore have a wavelength comparable to interatomic distances. With λ = 1 Å, we have:

$$E\ (\text{photon}) = hc/\lambda \approx 12.4\ \text{keV}.$$

In the soft x-ray/VUV region, on the other hand, the question tends to be "what are the electrons doing as they migrate between the atoms?" The emphasis turns to studies of chemical bonding and valence band structures using techniques such as photoemission and spectroscopy. It is now desirable that the photo*electron* should have a wavelength comparable to interatomic distances. With λ= 1 Å, we have

$$E\ (\text{electron}) = h^2 / 2\ m\lambda^2 \approx 150\ \text{eV}.$$

Photoemission investigations of valence bands and energy-momentum or E(**k**), relations are generally done using photon energies below 100 eV. Indeed, it is possible to scan the photoelectron wavevector out to the Brillouin zone boundary using photon energies as low as 20 eV.

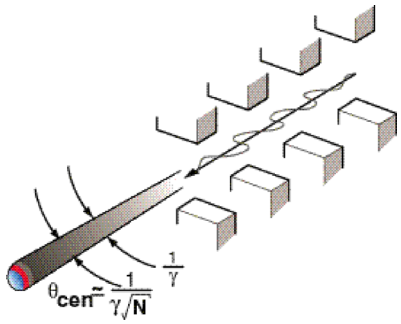
This natural separation into two energy regions is reflected in the establishment in the United States of two new synchrotron radiation facilities, the Advanced Photon Source (APS) at the Argonne National Laboratory optimized for delivery of hard x-rays, and the Advanced Light Source (ALS) at the Lawrence Berkeley National Laboratory optimized for soft x-ray/VUV science. These sources have been designed for high brightness.

Between the two extremes lies the "soft x-ray" region (100-1000 eV) which contains the binding energies of core levels of important elements, especially the first row of the periodic table. The binding energies of the 1s core level in C, N, and O are 290 eV, 400 eV, and 530 eV respectively. These elements have only one core level, and so core-level spectroscopy of these elements must be done the soft x-ray region. This region also contains the L-edges (2p core level to 3d valence level) of the transition metals. Particularly important are the L-edges of the elemental ferromagnets, Fe (710eV), Co (780eV), and Ni (850eV). In addition, the cuprate high-temperature superconductors and the manganate CMR (colossal magnetoresistance) materials have propelled the L-edges of Cu (930eV) and Mn (640eV) into prominence.

The intent of this article is to outline science being done using the soft x-ray and VUV part of the spectrum, with emphasis on the new opportunities offered by the high brightness of the ALS and its sister facilities elsewhere in the world. High brightness has three main advantages, and our examples have been chosen to illustrate these three advantages. For reasons of familiarity, the examples chosen will be taken from work done at the ALS and at the NSLS (National Synchrotron Light Source) at Brookhaven. Similar work is being done at the other sources, such as ELETTRA (Trieste), MAX-Lab (Lund), and BESSY-II (Berlin), and SRC (Madison), as well as facilities with moderately higher energy such as SSRL (Stanford).

The Brightness Advantage

The third generation synchrotron radiation sources have been designed for high brightness, sometimes called brilliance. The highest brightness is achieved with the use of "undulators", periodic magnetic structures that force the electrons in the synchrotron storage ring to oscillate about their central trajectory. The cumulative effect of many such undulations is to produce a beam of radiation in a very narrow angular cone about the forward direction. This, combined with the inherent smallness (30μm x 200μm) of the electron source in the ring, is the brightness advantage. For further reading, see D. Attwood, "Soft X-rays and Extreme Ultraviolet Radiation", Cambridge University Press (2000).



But what does brightness really buy you? Through a perfect optical system brightness is a conserved quantity which means that it is possible to put a lot of light into a small spot. This has three practical consequences:

- *Small spot on the sample.* Techniques (spectroscopy, diffraction, etc.) can now be performed with fine spatial resolution. With demagnifying mirrors, micron and submicron spot sizes can be achieved spawning a variety of x-ray microscopes. With use of Fresnel zone plates, spot sizes of 30nm are routine and 10nm should be possible.

- *Narrow slits.* With small spot size it is possible to cram a lot of light through very narrow slits of a monochromator or spectrometer. Brightness can therefore be traded for high resolving power, thereby driving a basic research program with much emphasis on strongly correlated electron systems in both the condensed state and gas phase.

- *Tiny pinholes*. It is likewise possible to cram a lot of light through tiny pinholes, thereby generating perfect spherical waves of high amplitude. This is the property of spatial coherence which can be exploited for interferometry, speckle and dynamic scattering experiments.

Spatial Resolution:
Wet and Dry Soft X-ray Microscopy

An important feature of the soft x-ray region is the "water-window", the photon energy range between the carbon K-edge (290eV) and the oxygen K-edge (530eV). In this range, organic matter (i.e. carbon) is absorbing whereas water (i.e. oxygen) is relatively transparent. This provides a contrast mechanism which permits microscopic analysis of cells in their natural aqueous environment. The richness of carbon near-edge spectra provides a chemical fingerprint, which serves as yet another contrast mechanism in the microscopic investigation of systems like polymer blends, which are of critical industry importance to the chemical industry. Let us focus first on an example of wet cell biology.

Confocal microscopy in the visible region is a powerful and widely used technique in cell biology. Its spatial resolution is limited by the wavelength of light. With use of Fresnel zone plates as focussing elements in the water window, it is possible to see features 5-10 times smaller than in the optical confocal microscope. Indeed the large potential impact of soft x-ray microscope on biology was one of the driving forces behind the building of low-energy third generation synchrotron radiation facilities. It is only recently, however, that this dream has started to come true.

A team of cell biologists working under the leadership of Carolyn Larabell is addressing the frontier problem of the localization, co-localization, and redistribution of proteins as they perform their functions in cells. Figure 1 shows mouse 3T3 fibroblasts taken with a spatial resolution of 25nm [1]. The sample was taken freshly from culture and cryofixed (i.e. frozen with liquid-nitrogen cooled helium gas) and, therefore, the cells more closely resemble their native state than do chemically fixed cells. Using this approach, the cellular ultrastructure is extremely well preserved and is revealed with a unique combination of high spatial resolution and good contrast. Numerous organelles, granules of various sizes, and tubular structures such as mitochondria are readily seen in the cytoplasm. Even the contents of the nucleus, which is approximately 5-μm thick in these cells, can be visualized without the need for physically sectioning the cell. Nuclear structures such as the nucleoli, which are densely packed with RNA, appear as dense bodies and the nuclear envelope, a double-layered membrane encircling the entire nucleus, appears as a distinct line encircling the nucleus. The striking contrast of the cellular ultrastructure is derived from imaging in the water window; these cells had not been exposed to any chemical fixatives or contrast enhancement agents. Due to the unique differential contrast provided by x-ray imaging, the location of specific proteins and nucleic acids is readily identified using gold-tagged labels that are easily distinguished from biological constituents [1]. Future developments will include collection of images at different tilt angles followed by tomographic reconstructions to restore the three-dimensional information. The use of X-ray microscopy is rapidly emerging as an important tool for determining structure-function relationships of proteins and cells. The future use of zone plate lenses with better resolution will enhance the information generated using this approach.

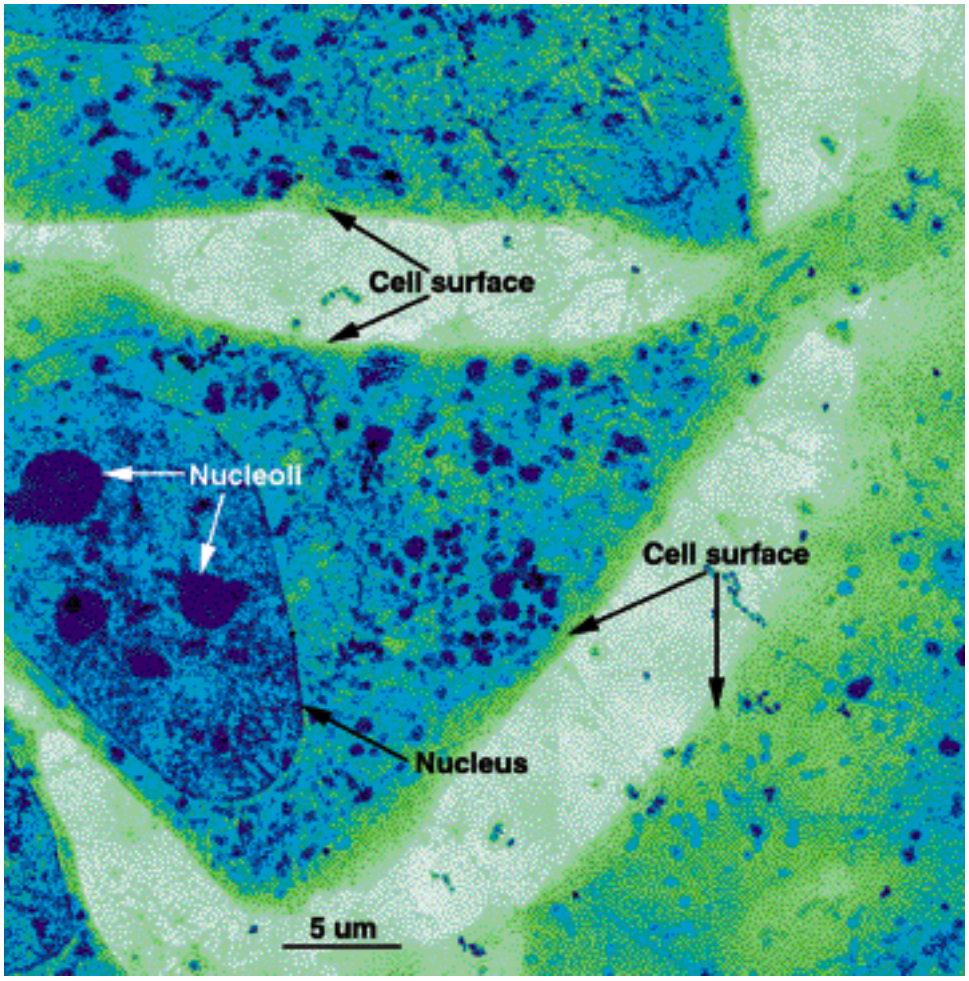


FIGURE 1. Soft x-ray transmission micrograph of mouse 3T3 fibroblasts. The data were taken in the "water window" with a spatial resolution of 30nm permitting observation of features such as nucleoli and the sharp nuclear membrane not resolvable with optical confocal microscopy. (Courtesy C. Larabell)

Fresnel zone plates represent only one of the ways to achieve fine spatial resolution. Another way is to use electron optics rather than photon optics. In photoelectron emission microscopy (PEEM) the principle is to illuminate an area on the sample and then to pass the ejected photoelectrons through an electron-microscope column to produce an enlarged image of the illuminated spot. Since the detected particles are electrons, the experiment must be done in vacuum thereby precluding the investigation of wet samples. Nevertheless, there are plenty of dry systems of interest.

Fritjhof Nolting and collaborators [2] have recently addressed the unresolved problem of exchange bias, i.e. the alignment of ferromagnetic spins by an antiferromagnet. This is something of an old chestnut in condensed matter physics, but has assumed considerable recent importance because of its relevance to the artificial magnetic layered structures used in the manufacture of devices such as magnetic read heads and magnetic memory cells. To build such a magnetic structure, it has proved necessary to use an antiferromagetic material as a substrate in order to pin the orientation of the first ferromagnetic layer. The mechanism is not well understood. By a very artful switch between two contrast mechanisms, linear dichroism for antiferromagnetic contrast and circular dichroism for ferromagnetic contrast, Nolting et al have provided what has hitherto been lacking, an incisive technique to study the problem.

The system chosen for study was thin ferromagnetic film of Co on an antiferromagnetic substrate of $LaFeO_3$. Figure 2 shows the PEEM micrographs and spectra. Linear dichroism (difference in the absorption of linearly polarized x-rays with the antiferromagntic axis parallel or perpendiclar to the electric vector of the light) at the Fe L-edge provides sufficient contrast to distinguish the vertically and horizontally oriented antiferromagnetic domains. Switching to circular dichroism (difference in absorption between left and right circularly polarized photons) at the Co L-edge, it can be seen that the ferromagnetic domains are indeed aligned with the domains of the antiferromagnetic substrate with the ferromagnetic magnetization pointing in either direction. It is then possible with PEEM to zoom in on specific areas and measure the magnetization reversal. This can be done as a function of temperature and, eventually, using the pulsed nature of synchrotron radiation, it will be done as a function of time. The present spatial resolution of PEEM is 20nm, but it is expected that aberration corrected instruments being developed in the U.S. and Germany will reduce this to 2nm.

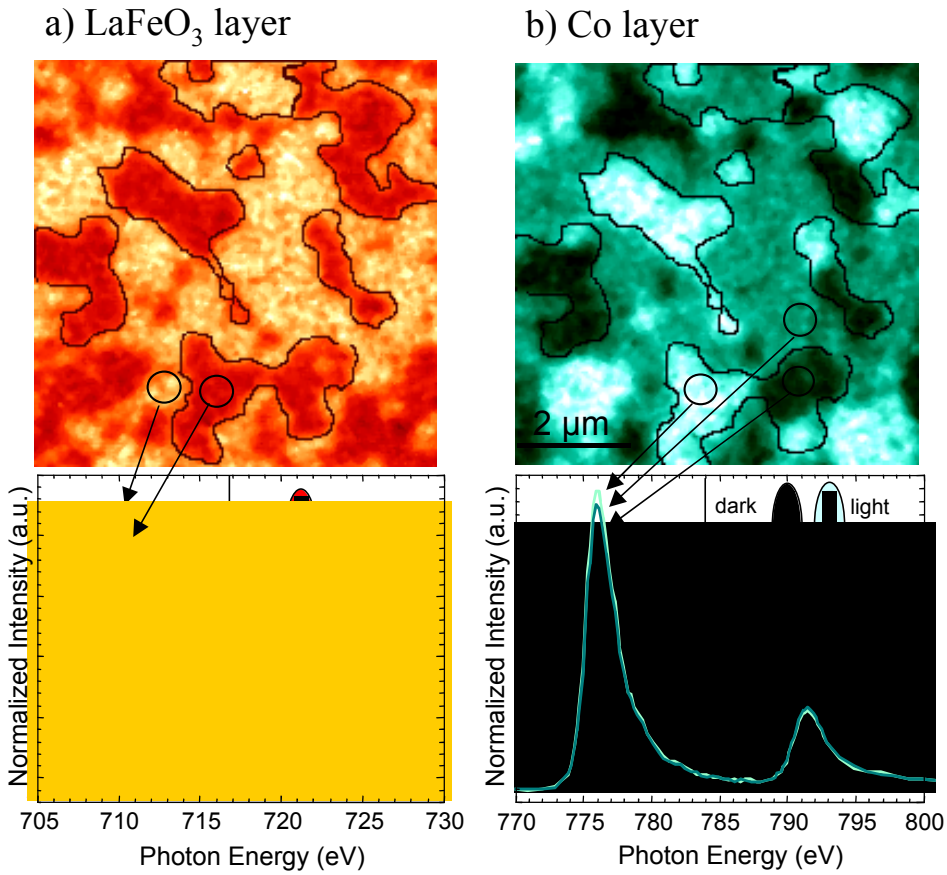


FIGURE 2. Images and local spectra from a ferromagnetic Co layer on an antiferromagnetic $LaFeO_3$ substrate. (a) Antiferromagnetic contract is obtained at the Fe L-edge using linear dichroism, the difference in absorption between parallel and perpendicular orientation of the antiferromagnetic axis and the electric vector of the light. (b) Ferromagnetic contrast at the Co L-edge using circular dichroism, the difference in absorption between left and right circular polarization. Note that the ferromagnetic Co domains align with the substrate antiferromagnetic domains, but can split into subdomains of opposite magnetization. (Courtesy S. Anders.)

Spectral Resolving Power:
Electronic Structure of High Tc Superconductors

One of the most tantalizing problems in condensed-matter physics is the origin of high temperature superconductivity. Phil Anderson has declared [4] that angle-resolved photoemission spectroscopy (ARPES) will provide the smoking gun and "is, for this problem, the experiment that will play the role that tunneling played for BCS." Great strides have indeed been made. ARPES experiments have revealed the d-wave nature of the superconductive coupling mechanism [5] and the existence of a "pseudogap". Attention has turned recently to the existence of stripes [6] and we discuss here some ARPES experiments by Zhi-xun Shen and co-workers [7] which make a connection between stripes and the Fermi surface, i.e. the boundary in k-space separating the occupied and unoccupied electronic states.

The experiments were done on a model cuprate compound ($La_{1.28}Nd_{0.6}Sr_{0.12}$) CuO_4 (Nd-LSCO). This remarkable compound has the one-eighth ($Sr_{0.12}$) strontium doping at which superconductivity is actually supressed. The replacement of some La with Nd serves to stabilize the stripe phase which consists, at this doping, of rows of charge carriers separate by triple rows of antiferromagnetically ordered spins whose ordering flips phase across the charge stripes. (See Figure 3). Shen et al extract a Fermi surface by integrating the photoemission spectra over an energy range near the Fermi level to obtain electron momentum density maps that reveal the Fermi surface information. The Fermi surface displays the shape of a cross, although it is thought that this is the superposition of two bars corresponding to domains having the two alternative orientations of the stripes. The boundaries of the bars are given by $|k_x| \leq \pi/4$ and $|k_y| \leq \pi/4$ implying that the electron system is highly one-dimensional consistent with the fourfold periodicity and the one-eighth doping. The data are inconsistent with the more rounded Fermi surface based on the two-dimensional copper-oxygen planes.

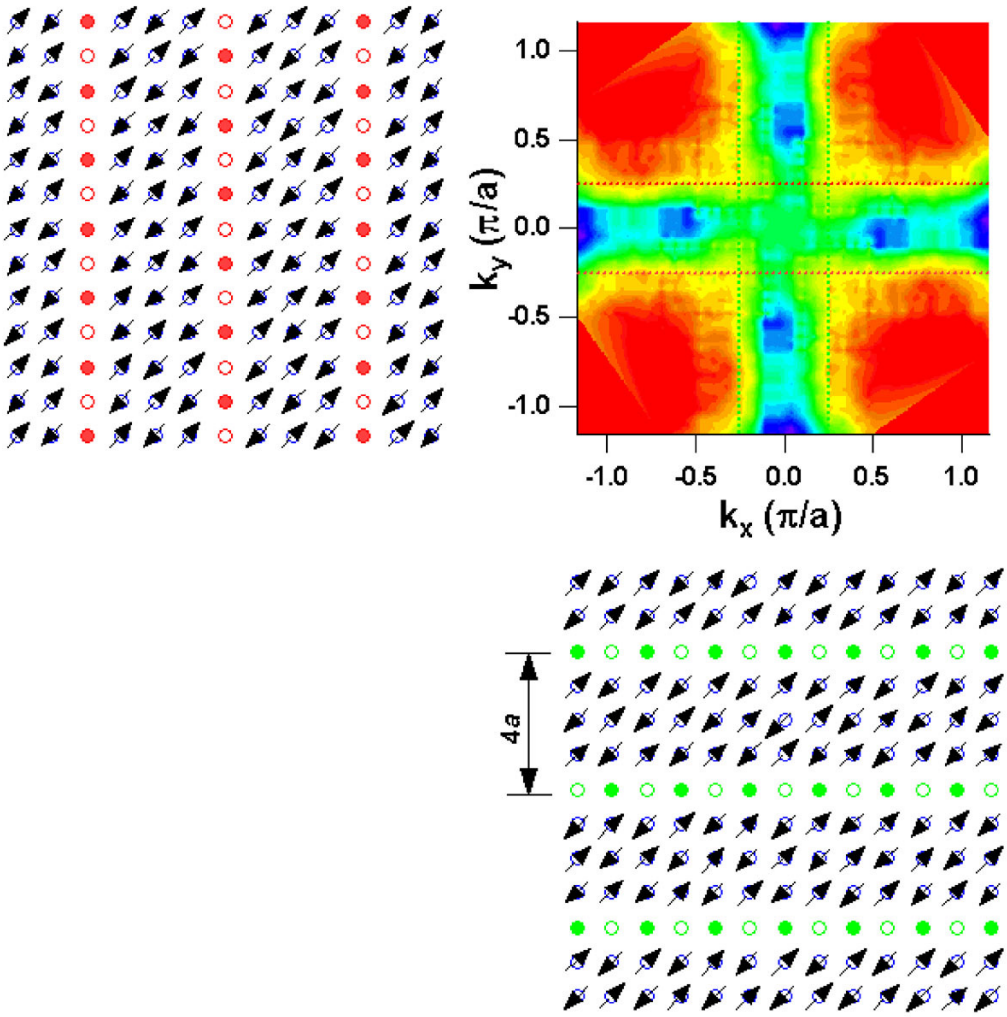


FIGURE 3. Fermiology meets stripology. In the stabilized stripe phase, rows of charge carrying atoms are separated by triple-rows of antiferromagnetic material with a π phase slip across the charge carrying rows. The "Maltese Cross" Fermi surface (upper right) is confined within $| k_x | \leq \pi /4$ and $| k_y | \leq \pi/4$ consistent with a superposition of the two alternative domain orientations of the stripe phase.

High resolving power is permitting photoemission spectroscopists to approach ever closer to the Fermi level, E_F. Energy band structures ($E(\mathbf{k})$ relations) can now be measured with great precision. For example, the long anticipated kink in the $E(\mathbf{k})$ relation close to E_F due to the electron-phonon interaction has now been observed by a number of groups. Attention has turned also to the lifetime width of peaks in the photoemission spectra. The energy dependence of the lifetime width in the vicinity of E_F is a strong indicator of Fermi-liquid or non-Fermi-liquid like behavior.

The measured photoelectron energy spectrum at a given angle of emission is proportional to the spectral function given by

$$A(\mathbf{k},\omega) \alpha \frac{\mathrm{Im}\,\Sigma(\mathbf{k},\omega)}{[\omega - E_k - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

where $\mathrm{Re}\Sigma(\mathbf{k},\omega)$ and $\mathrm{Im}\Sigma(\mathbf{k},\omega)$ are the real and imaginary parts of the self energy. Modern state-of-the-art photoemission spectrometers display in parallel two dimensions of information, photoelectron energy and angle. Peter Johnson and his collaborators, working at the Brookhaven National Synchrotron Light Source, have shown that the cleanest cut through this space for the determination of the $\mathrm{Im}\Sigma$ is the momentum distribution curve (MDC) which is the photoemission intensity as a function of momentum k at constant excitation energy ω. The MDC's are immune from the distorting effects of the Fermi function and the inelastic background to the spectra.

Figure 4 shows a comparison of lifetime widths measured by Johnson's group on optimally doped $Bi_2\,Sr_2\,CaCu_2\,O_{8+\delta}$ (Bi2212) with those measured on testbed system, a molybdenum surface state [8]. Since the measurement of photoemission line widths is fraught with experimental artifacts, it is highly desirable to test the method on a well-understood system. A well established surface state on the Mo(110) surface provides such a test bed system. The ω dependence of the $\mathrm{Im}\Sigma$ is well described by a contribution due to the electron-phonon interaction plus a ω^2 term characteristic of a Landau Fermi liquid. By contrast, the measurements of $\mathrm{Im}\Sigma$ for Bi2212 are clearly inconsistent with Fermi-liquid behavior. First, the data lie above the curve $\omega = |2\mathrm{Im}\Sigma|$ whereas the validity of Fermi liquid theory requires $|2\mathrm{Im}\Sigma| < \omega$. Second, the ω dependence and T dependence of the self energy suggest that the system exhibits quantum critical behavior. This interpretation, as well as the discussion above concerning the stripe phase, remains controversial. The important conclusion for the purpose of this article is that the incisiveness of ARPES does indeed look likely to provide Anderson's smoking gun, and that the work requires high brightness synchrotron radiation in the vacuum ultraviolet region.

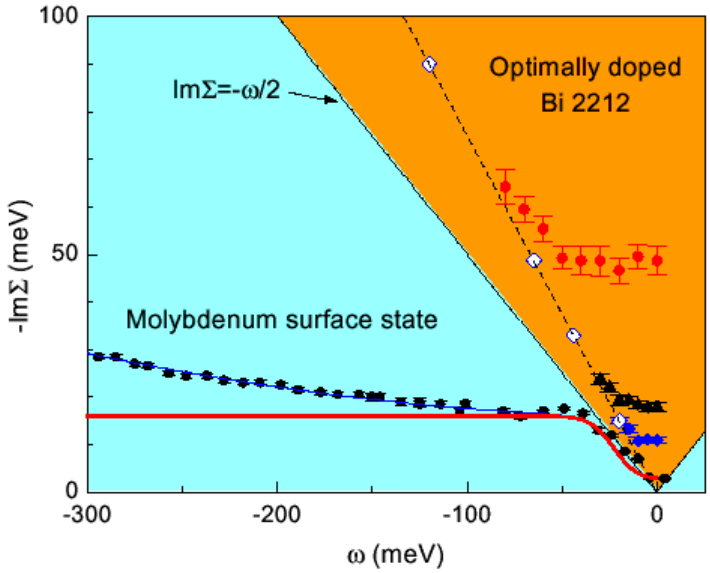


FIGURE 4. Lifetime widths on approaching the Fermi level as measured by angle-resolved photoemission spectroscopy. The testbed results on a Mo(110) surface state (black circles) are well understood in terms of Fermi-liquid theory (blue parabola) and electron phonon interaction (red curve). This is in contrast with results on the superconducting material Bi2212. The red cicles, black triangles and blue diamonds correspond to lifetime widths at temperatures 300K, 90K and 48K respectively and lie in a region clearly inconsistent with Fermi liquid behavior. (Courtesy T. Valla)

Coherence :
Interferometry for EUV Lithography

According to Moore's famous Law, the density of circuit elements on microchips has doubled roughly every two years, resulting in smaller, faster, and cheaper computers. The present technology, optical lithography, cannot continue indefinitely on this course. The materials from which one could conceivably make lenses, CaF_2, MgF_2 and LiF will not transmit light at wavelengths less than 100nm. One of the options being considered is to switch from refractive optics to reflective optics, i.e. mirrors. The availability of Mo/Si mirror coatings having reflectances as high as 70% provides special impetus for this technology The optimum wavelength determined as a compromise between the conflicting requirements of fine lateral spatial resolution and large depth of focus into the chip structure, is about 13nm. This wavelength is popularly referred to as "extreme ultraviolet" or EUV. The corresponding photon energy is 100eV which qualifies as "soft x-ray."

A consortium of microelectronics companies (Intel, Motorola, Advanced Micro Devices) has joined forces with a consortium of National Laboratories (Livermore, Sandia and Berkeley Labs) to build a prototype EUV stepper, an optical camera of the sort to produce small computer chips. A schematic of such a stepper is shown in Figure 5. It comprises a four-mirror optic which produces a demagnified image of the mask on the wafer. The mirrors must be curved and coated with interfering multilayers in order to generate a high reflectivity at 13nm. The EUV source in actual chip manufacture will not be synchrotron radiation but will be derived from an EUV emitting plasma. The role of synchrotron radiation for this technology is optics metrology.

At Berkeley Lab, Jeff Bokor is leading a team that has been entrusted with the task of interferometric characterization of the required high precision mirrors. An old adage says "if you can't measure it, you can't make it." The team has developed an at-wavelength phase-shifting point-diffraction interferometer (PS/PDI) to meet the requisite specifications [9]. The principle of the PS/PDI is to first pass the beam from an ALS undulator through a pinhole to generate a high amplitude perfect spherical wave. A diffraction grating then splits the light into an order (the test beam) which passes through the test optic generating an aberrated wavefront characteristic of the imperfections in the test optic. Another order passes unobstructed through the optic but then is forced through a second pinhole to generate a perfectly spherical reference wavefront which interferes with the test beam. The resulting interferogram reveals the departures from figure accuracy. The design goal was for a test accuracy of 0.10nm over the surface of the mirror. The accuracy actually achieved is 0.05nm. This corresponds to the Bohr radius of a hydrogen atom! A four-mirror optic manufactured at Sandia has already been tested with very satisfactory results. This work demonstrates that coated optics to the desired tolerance can be produced, thereby enhancing the prospects of EUV lithography as the next technology of choice for the manufacture of ever denser microchips.

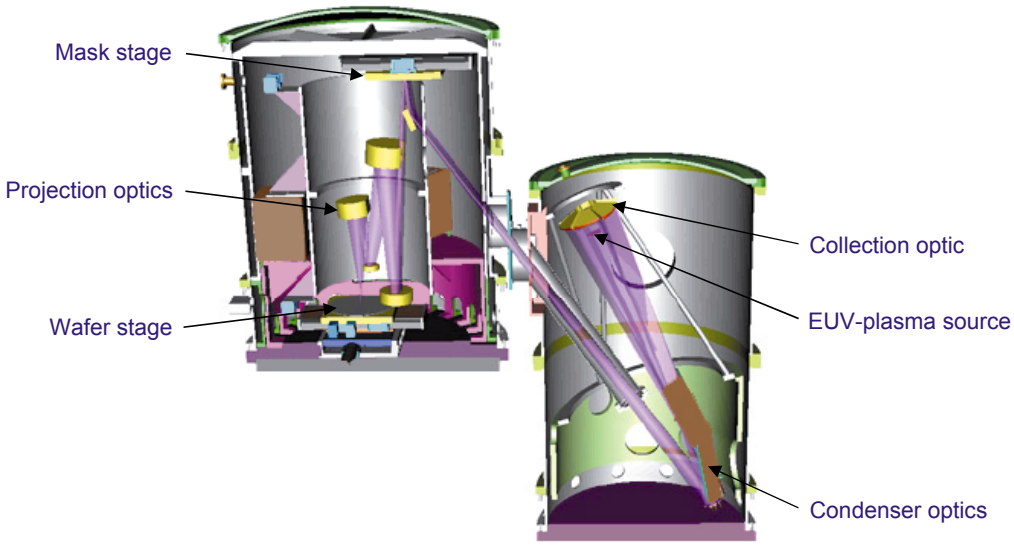


FIGURE 5. Schematic representation of an optical system for the manufacture of microchips using EUV projection lithography at 13nm. Multilayer coated mirrors in the stepper create a demagnified image of the mask onto the wafer. A plasma source provides the EUV radiation. (Courtesy R. Stulen, Sandia National Laboratory)

References

1. C. A. Larabell, D. Yager, and W. Meyer-Ilse, in *X-ray Microscopy,Proceedings of the Sixth International Conference,* Berkeley, CA 2-6 August 1999, edited by W.Meyer-Ilse, T. Warwick and D. Attwood, American Institute of Physics, 2000, p. 107.

2. C. Jacobsen, S. Abend, T. Beetz, M. Carlucci-Dayton, M. Feser, K. Kaznacheyev, J. Kirz, J. Maser, U. Neuhausler, A. Osanna, A. Stein, C. Vaa, Y. Wang, B. Winn, S. Wirick, in Ref 1, p.12. D.Weiss, G. Schneider, B. Niemann, P. Guttmann, D. Rudolph, and G. Schmahl, in Ref. 1, p.123.

3. F. Nolting, A. Scholl, J. Stohr, J. Fompeyrine, H. Siegwart, J.-P. Locquet, S.Anders, J. Lüning, E.E. Fullerton, M. F. Toney, M. R. Scheinfein, and H. A. Padmore, Nature, **405**, 767 (2000).

4. P.W. Anderson, Physics Today, **44** (6), 60 (1991).

5. Z. -X. Shen, D. S. Dessau, B. O. Wells, D. M. King, W. E. Spicer, A. J. Arko, D. Marshall, L.W. Lombardo, A. Kapitulnik, P. Dickinson, S. Doniach, J. DiCarlo, T. Loeser, and C. H. Park, Phys. Rev. Lett. **70**, 1553 (1993).

6. J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura, and S. Uchida, Nature, **375**, 561 (1995).

7. Z.J. Zhou, P. Bogdanov, S.A. Kellar, T.Noda, H. Eisaki, S. Uchida, Z.Hussain and Z.-X. Shen, Science, **286**, 189 (1999).

8. T. Valla, A.V. Federov, P.D. Johnson. B.O. Wells, S.L. Hulbert, Q. Li, G.D. Gu and N. Koshizuka, Science, **285**, 2110 (1999); T. Valla, A.V. Federov, P.D. Johnson and S. L. Hulbert, Phys. Rev. Lett, **83**, 2085 (1999).

9. P. Naulleau, K.A. Goldberg, S. Lee, C. Chang, C. Bresloff, P. Batson, D. Attwood and J. Bokor, SPIE **331**, 114 (1998); G. W. Gwyn, R. Stulen, D. Sweeney and D. Attwood, J. Vac. Sci. Tech, **B16**, 3142 (1998).

Affiliation

Neville Smith is Division Deputy for Science at the Advanced Light Source, Lawrence Berkeley National Laboratory.